

Appendix A

This appendix contains a summary of the experimental data obtained using methane (99.99%) primarily in water prepared by reverse osmosis (RO water) or artificial seawater made from Instant Ocean® to a salinity of 35 as determined by a PINPOINT® salinity monitor calibrated against a 53 mS calibration fluid, both from American Marine, Inc. The experimental data are ordered by the date the experiments were performed. These data primarily include observations of individual bubbles, however, small bubble swarms were also observed. The data are grouped into sets. In data sets 1 through 5 the dissolved CH₄ concentration was incrementally increased to promote hydrate formation. Data Set 6 consistent of 17 consecutive bubble with the only dissolved gas coming from the dissolution of the bubbles. Data Set 7 was performed with seawater. A new data set was started when the water in the HWTF was drained and degassed.

This Appendix is divided into sections representing each data set. The experimental data are shown in tabular and graphical form. Comments and notes are also included to facilitate understanding and interpretation of the results.

Table A-0 explains the column headings used in the following tables that present the various data sets.

Table A-0. Explanation of column headings in data tables.

Column Heading	Explanation
Obser. #	An index number used for reference purposes and graphing the data.
Sortable Date	When the observation was made.
Bubble #	What was observed; e.g., a bubble (and its order if needed), a stream or swarm of bubbles, or gas exsolution or heating events to eliminate any hydrate memory effect.
Avg. P (MPa _{gauge})	Pressure in the HWTF during the observation, either from logbook entries or in the case of dissolution measurements, the average from the data processed to obtain the dissolution rate.
Avg. T (°C)	Temperature in center of the viewing section of the HWTF obtained in the same way as the pressure.
Observ. Time (min)	Time the bubble was in view or duration of another event. The time interval over which dissolution rate measurements were made can be different (see Appendix C).
X _{exp} (mol frac.)	Dissolved gas concentration in the experiment when the observation was made. Expressed as mole fraction of methane in water or seawater.
X _{LH} (mol frac.)	Calculated liquid/hydrate (LH) equilibrium conditions during the observation. The minimum dissolved gas concentration for hydrate stability. A thermodynamic model developed on this project from first principles was used to obtain these values.
(X _{exp} - X _{hyd})/X _{exp}	Ratio reflecting relative gas saturation. -1.0 indicates no dissolved gas. 0 indicates the system is at LH equilibrium. Values > 0 indicate supersaturation.
P _{VLH} @ avg T (MPa _{absolute})	The vapor/liquid/hydrate equilibrium pressure at the average experimental temperature.
P _{avg} / P _{VLH}	The ratio of the average pressure to VLH pressure. Values ≥ 1 are within the region of hydrate stability.
Dissolution rate (um/s)	The dissolution rate of the bubble.

The key to Table A-1 applies to all tables in this appendix. The caption to Figure A-1 applies to all figures in this appendix.

Table A-1. CH4 Data Set 1

Data Set A-1: CH4 in RO water. 1 to 3°C. Performed from 5/8/2012 to 05/22/2012.											
Obsr. #	Sortable Date	Bubble #	Avg. P (MPa _{gauge})	Avg T (°C)	Observ. Time (min)	X _{exp} (mol frac.)	X _{LH} (mol frac.)	(X _{exp} - X _{LH})/X _{LH}	P _{VLH} @ avg T (MPa _{absolute})	P _{avg} / P _{VLH} (MPa _{absolute})	Dissolution rate (um/s)
1	5/8/2012	1st Bubble	20.022	1.7	34	0.000003	0.001132	-0.997350	3.087	6.518	0.963
2	5/8/2012	PIV stream	19.995	1.6	2	0.000053	0.001126	-0.952931	3.059	6.570	
3	5/8/2012	Gas Addition	19.995	1.6	54	0.000464	0.001126	-0.587922	3.059	6.570	
4	5/8/2012	Exsolution	2.875	1.7	7				3.087	0.964	
5	5/8/2012	Increased P	20.098	1.8	18	0.000464	0.001138	-0.592267	3.116	6.483	
6	5/8/2012	2nd Bubble	20.043	1.8	16	0.000467	0.001138	-0.589631	3.116	6.465	
7	5/9/2012	1st Bubble	20.043	1.5	20	0.000470	0.001119	-0.579982	3.031	6.647	0.769
8	5/9/2012	1st Bubble	24.794	1.5	6	0.000470	0.001095	-0.570776	3.031	8.214	1.133
9	5/9/2012	1st Bubble	30.068	1.6	26	0.000470	0.001075	-0.562791	3.059	9.863	0.746
10	5/10/2012	1st Bubble	9.984	1.2	35	0.000540	0.001115	-0.530435	2.946	3.423	1.026
11	5/10/2012	2nd Bubble	19.981	1.1	28	0.000541	0.001094	-0.505484	2.918	6.883	0.750
12	5/10/2012	3rd Bubble	29.999	1.1	51	0.000542	0.001045	-0.481340	2.918	10.317	0.680
13	5/11/2012	1st Bubble	29.985	0.6	81	0.000702	0.001016	-0.309055	2.773	10.850	0.666
14	5/14/2012	1st Bubble	29.903	1.3	72	0.000843	0.001058	-0.203214	2.974	10.088	0.648
15	5/15/2012	1st Bubble	9.991	1.9	36	0.000997	0.001198	-0.167780	3.144	3.209	0.824
16	5/15/2012	2nd Bubble	20.043	1.8	43	0.000999	0.001139	-0.122915	3.116	6.465	0.646
17	5/15/2012	3rd Bubble	30.034	1.5	68	0.001001	0.001070	-0.064486	3.031	9.944	0.611
18	5/16/2012	1st Bubble	9.984	1.7	66	0.001138	0.001184	-0.038851	3.087	3.267	0.755
19	5/16/2012	2nd Bubble	29.917	1.2	40	0.001140	0.001049	0.086749	2.946	10.190	-0.035
20	5/16/2012	2nd Bubble	20.133	1.3	22	0.001140	0.001106	0.030741	2.974	6.803	-0.008
21	5/16/2012	2nd Bubble	10.170	1.3	21	0.001140	0.001156	-0.013841	2.974	3.453	
22	5/16/2012	2nd Bubble	5.164	1.3	3	0.001140	0.001181	-0.034716	2.974	1.770	
23	5/18/2012	1st Bubble	9.970	1.9	19	0.001141	0.001198	-0.047579	3.144	3.203	
24	5/18/2012	1st Bubble	4.130	2	5	0.001141	0.001237	-0.077607	3.173	1.333	
25	5/18/2012	1st Bubble	4.530	2.1	16	0.001141	0.001242	-0.081320	3.202	1.446	
26	5/18/2012	1st Bubble	4.530	2.3	29	0.001141	0.001257	-0.092283	3.261	1.420	0.566
27	5/18/2012	1st Bubble	4.516	2.5	18	0.001141	0.001272	-0.102987	3.322	1.390	0.292
28	5/18/2012	1st Bubble	4.420	2.5	1	0.001141	0.001273	-0.103692	3.322	1.361	
29	5/18/2012	1st Bubble	4.344	2.5	2	0.001141	0.001273	-0.103692	3.322	1.338	
30	5/18/2012	1st Bubble	4.137	2.6	2	0.001141	0.001282	-0.109984	3.352	1.264	
31	5/18/2012	1st Bubble	3.951	2.5	17	0.001141	0.001275	-0.105098	3.322	1.220	
32	5/18/2012	1st Bubble	3.744	2.4	10	0.001141	0.001269	-0.100867	3.291	1.168	
33	5/18/2012	1st Bubble	4.502	2.1	129	0.001141	0.001242	-0.081320	3.202	1.438	
34	5/21/2012	Exsolution	3.144	2.8	5				3.415	0.950	
35	5/22/2012	1st Bubble	9.997	3.4	0.9	0.001142	0.001310	-0.128244	3.614	2.794	
36	5/22/2012	1st Bubble	9.997	3.4	0.1	0.001142	0.001310	-0.128244	3.614	2.794	
37	5/22/2012	2nd Bubble	9.997	3.4	6.0	0.001143	0.001310	-0.127481	3.614	2.794	
38	5/22/2012	PIV stream	19.995	3.4	109.0	0.001617	0.001250	0.293600	3.614	5.561	

Refer to Table A-0 for description of column headers.

Grey shading: In Observ. # column indicates a new bubble.**No shading:** In Observ. # column indicates observation of same bubble at different conditions.**Orange shading:** Indicates hydrate present on bubble or in HWTF.**Light orange shading:** In Observ. Time column indicates nothing came into view during this observation period.**Violet shading:** Indicates attempt to destroy hydrate memory effect by gas exsolution and/or heating as indicated.

(NOTE: Not all colors are needed in every table.)

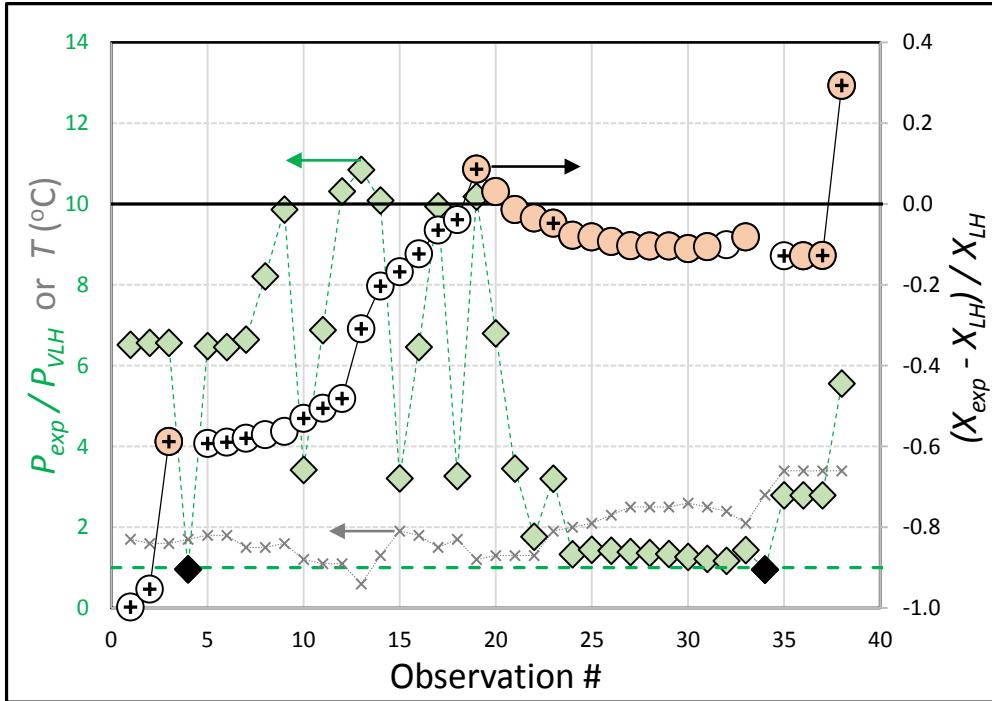


Figure A-1. Graphical representation of the sequence of observations in Data Set A-1 (CH4 in RO water).

Diamonds: Green represent P_{avg} / P_{VLH} . Black represent gas exsolution. White represents heating. Black with a white diamond in center represent gas exsolution with heating. Exsolution was used in attempts to eliminate a hydrate memory effect from prior hydrate formation in the system. Heating was also used for this purpose or to move the system out of a region were the gas was supersaturated with the dissolved gas. Not all exsolution/heating symbols are used in every graph.

Circles: White represent bubble or other observations indexed to $(X_{exp} - X_{LH}) / X_{LH}$, the relative gas saturation expressed as a fraction percent. Orange circles indicate hydrate present. A plus sign (+) in a circle indicates a new bubble released in the HWTF.

Crosses (x): Indicate average temperature.

Horizontal lines: Dashed green line indicates $P_{avg} / P_{VLH} = 1.0$ (i.e., $P_{avg} = P_{VLH}$). $P_{avg} / P_{VLH} > 1$ indicate the system is inside the region of thermodynamic hydrate stability with respect to VLH equilibrium. Solid black line indicates $X_{exp} = X_{LH}$, i.e., the experimental dissolved gas saturation equals the predicted gas saturation. Values > 0 indicates supersaturation. A value of -1.0 indicates no dissolved gas present in the aqueous phase.

Notes on Data Set A-1.

Observation #3 represents the first gas addition to increase the dissolved CH4 content ((X_{exp})), which was performed well inside the VLH region of stability ($P_{avg} / P_{VLH} = 6.570$; see Table A-1 for more detail). Hydrate was noted after injection when the HWTF was mixed by changing flow direction. A short exsolution of gas was performed by dropping the pressure until the system was at $P_{avg} / P_{VLH} = 0.964$, or just outside of the VLH region of hydrate stability. The absence of hydrate on the next 12 bubbles and gas additions at lower pressure, the latter made after Observations #9, #12, #13, #14, and #17, indicates that mixing in the HWTF well inside the VLH region for hydrate stability was the cause of hydrate formation in #3 rather than bulk dissolved gas concentration levels or bubble surface properties.

Table A-2. CH4 Data Set 2

Data Set A-2: CH4 in RO water. 1 to 3°C. Performed from 5/31/2012 to 06/13/2012.												
Observ. #	Sortable Date	Bubble #	Avg. P (MPa _{gauge})	Avg T (°C)	Observ. Time (min)	X _{exp} (mol frac.)	X _{LH} (mol frac.)	(X _{exp} - X _{LH}) / X _{LH}	P _{VLH} @avg T (MPa _{absolute})	P _{avg} / P _{VLH} (MPa _{absolute})	Dissolution rate (um/s)	
1	5/31/2012	1st bubble	10.004	4.7	31	0.000002	0.001417	-0.998589	4.118	2.454	1.482	
2	6/1/2012	1st bubble	20.078	4.6	9	0.000006	0.001343	-0.995532	4.075	4.952	1.771	
3	6/1/2012	2nd bubble	19.974	4.8	47	0.000008	0.001360	-0.994117	4.161	4.825	1.031	
4	6/1/2012	3rd bubble	29.979	4.9	69	0.000012	0.001304	-0.990799	4.205	7.153	0.920	
5	6/4/2012	1st bubble	10.004	4.5	74	0.001058	0.001400	-0.244149	4.033	2.506	0.918	
6	6/4/2012	2nd bubble	20.036	4.5	26	0.001060	0.001335	-0.206063	4.033	4.993	0.754	
7	6/4/2012	3rd bubble	29.992	4.4	40	0.001062	0.001266	-0.161078	3.992	7.539	0.729	
8	6/5/2012	1st bubble	10.025	4.5	44	0.001274	0.001400	-0.089747	4.033	2.511	0.829	
9	6/5/2012	2nd bubble	19.995	4.4	59	0.001276	0.001327	-0.038686	3.992	5.035	0.713	
10	6/6/2012	1st bubble	29.972	4.4	43	0.001278	0.001266	0.009451	3.992	7.534	0.690	
11	6/6/2012	2nd bubble	29.999	4.4	38	0.001280	0.001266	0.011164	3.992	7.541	0.683	
12	6/6/2012	3rd bubble	30.006	4.3	37	0.001282	0.001258	0.018802	3.951	7.620	0.678	
13	6/7/2012	1st bubble	9.970	4.3	54	0.001433	0.001383	0.036153	3.951	2.549		
14	6/7/2012	2nd bubble	19.967	4.5	38	0.001435	0.001335	0.074906	4.033	4.976		
15	6/7/2012	3rd bubble	30.048	4.5	36	0.001437	0.001273	0.128830	4.033	7.476		
16	6/8/2012	1st bubble	9.984	4.7	67	0.001485	0.001417	0.047879	4.118	2.449	0.740	
17	6/8/2012	2nd bubble	19.988	4.6	42	0.001486	0.001344	0.106055	4.075	4.930	0.666	
18	6/8/2012	3rd bubble	30.041	4.4	45	0.001488	0.001266	0.175355	3.992	7.551	-0.035	
19	6/8/2012	3rd bubble	27.600	4.5	8	0.001488	0.001288	0.155280	4.033	6.869	-0.236	
20	6/8/2012	3rd bubble	24.235	4.5	6	0.001488	0.001309	0.136746	4.033	6.034	0.054	
21	6/8/2012	3rd bubble	20.712	4.5	21	0.001488	0.001331	0.117956	4.033	5.161	-0.168	
22	6/8/2012	3rd bubble	17.244	4.6	9	0.001488	0.001361	0.093314	4.075	4.257		
23	6/8/2012	3rd bubble	13.803	4.6	14	0.001488	0.001383	0.075922	4.075	3.412		
24	6/8/2012	3rd bubble	10.363	4.6	8	0.001488	0.001406	0.058321	4.075	2.568		
25	6/8/2012	3rd bubble	6.902	4.4	10	0.001488	0.001412	0.053824	3.992	1.754		
26	6/8/2012	3rd bubble	4.371	4.5	9	0.001488	0.001437	0.035491	4.033	1.109		
27	6/8/2012	Exsolution	3.806	4.6	25				4.075	0.959		
28	6/11/2012	1st bubble	10.032	4.4	2	0.001490	0.001391	0.071172	3.992	2.539		
29	6/11/2012	1st bubble	10.032	4.5	3	0.001490	0.001400	0.064286	4.033	2.513		
30	6/11/2012	2nd bubble	6.895	4.5	0.45	0.001491	0.001420	0.050000	4.033	1.735		
31	6/11/2012	2nd bubble A	7.743	4.7	9	0.001491	0.001433	0.040475	4.118	1.905	0.754	
32	6/11/2012	2nd bubble B(D)	8.425	4.8	9	0.001491	0.001437	0.037578	4.161	2.049	0.622	
33	6/11/2012	2nd bubble C	9.101	4.8	6	0.001491	0.001432	0.041201	4.161	2.212	0.118	
34	6/11/2012	2nd bubble D(B)	8.405	4.9	10	0.001491	0.001445	0.031834	4.205	2.023	0.661	
35	6/11/2012	2nd bubble E	7.708	4.9	1	0.001491	0.001450	0.028276	4.205	1.857		
36	6/11/2012	3rd Bubble	6.895	4.9	0.5	0.001492	0.001456	0.024725	4.205	1.664		
37	6/11/2012	3rd bubble A	7.681	5	12	0.001492	0.001459	0.022618	4.250	1.831	0.306	
38	6/11/2012	3rd bubble B	7.343	5.2	12	0.001492	0.001480	0.008108	4.341	1.715	0.402	
39	6/11/2012	3rd bubble C	6.998	5.3	10	0.001492	0.001492	0.000000	4.388	1.618	0.243	
40	6/11/2012	3rd bubble D	6.660	5.4	5	0.001492	0.001503	-0.007319	4.435	1.524	-0.087	
41	6/11/2012	3rd bubble E	6.309	5.4	5	0.001492	0.001506	-0.009296	4.435	1.445	0.695	
42	6/11/2012	3rd bubble F	5.964	5.5	8	0.001492	0.001518	-0.017128	4.484	1.353	0.550	
43	6/11/2012	3rd bubble	5.612	5.6	5	0.001492	0.001530	-0.024837	4.533	1.261		
44	6/11/2012	3rd bubble G	5.295	5.6	5	0.001492	0.001532	-0.026110	4.533	1.191	0.000	
45	6/11/2012	3rd bubble H	4.957	5.7	3	0.001492	0.001544	-0.033679	4.582	1.104	1.334	
46	6/11/2012	3rd bubble	5.861	5.7	1	0.001492	0.001537	-0.029278	4.582	1.301		
47	6/11/2012	3rd bubble I	6.329	5.7	2	0.001492	0.001534	-0.027379	4.582	1.403	-0.197	
48	6/11/2012	3rd bubble J	4.971	5.7	2	0.001492	0.001544	-0.033679	4.582	1.107	1.255	
49	6/11/2012	3rd bubble	6.205	5.7	2	0.001492	0.001535	-0.028013	4.582	1.376		
50	6/11/2012	Exsolution	4.275	5.5	944				4.484	0.976		

Continued on next page.

Data Set A-2 continued: CH4 in RO water. 1 to 3°C. Performed from 5/31/2012 to 06/13/2012.												
Observ. #	Sortable Date	Bubble #	Avg. P (MPa _{gauge})	Avg T (°C)	Observ. Time (min)	X _{exp} (mol frac.)	X _{LH} (mol frac.)	(X _{exp} - X _{LH}) / X _{LH}	P _{VLH} @ avg T (MPa _{absolute})	P _{avg} / P _{VLH} (MPa _{absolute})	Dissolution rate (um/s)	
51	6/12/2012	1st bubble	7.584	5.6	0.1	0.001493	0.001515	-0.014521	4.533	1.696		
52	6/12/2012	2nd bubble	7.736	5.8	14	0.001495	0.001534	-0.025424	4.633	1.692	0.654	
53	6/12/2012	3rd bubble	10.011	6.1	53	0.001496	0.001546	-0.032031	4.788	2.112	0.755	
54	6/12/2012	4th bubble	19.995	6.1	31	0.001497	0.001472	0.016750	4.788	4.197	0.686	
55	6/12/2012	5th bubble A	30.041	6.2	6	0.001499	0.001411	0.062367	4.841	6.227	0.516	
56	6/12/2012	5th bubble	29.999	6.2	10	0.001499	0.001411	0.062367	4.841	6.218		
57	6/12/2012	5th bubble	5.454	6.3	16	0.001499	0.001600	-0.063125	4.895	1.135		
58	6/12/2012	5th bubble	5.081	6.3	1	0.001499	0.001603	-0.064878	4.895	1.059		
59	6/12/2012	5th bubble B	5.799	6.3	7	0.001499	0.001597	-0.061365	4.895	1.205	0.375	
60	6/12/2012	5th bubble	10.170	6.4	9	0.001499	0.001574	-0.047649	4.949	2.075		
61	6/12/2012	5th bubble	4.826	6.4	2	0.001499	0.001600	-0.063125	4.949	0.996		
62	6/12/2012	Exsolution	4.309	6.4	977					4.949	0.891	
63	6/13/2012	1st bubble	7.715	5.6	50	0.001500	0.001515	-0.009901	4.533	1.725	0.676	
64	6/13/2012	2nd bubble	7.715	5.5	2	0.001502	0.001505	-0.001993	4.484	1.743		
65	6/13/2012	3rd bubble	10.025	5.4	38	0.001503	0.001480	0.015541	4.435	2.283		
66	6/13/2012	4th bubble	19.974	5.4	6	0.001504	0.001410	0.066667	4.435	4.526		
67	6/13/2012	4th bubble	19.967	5.4	22	0.001504	0.001410	0.066667	4.435	4.525		
68	6/13/2012	5th bubble	29.999	5.4	1.8	0.001506	0.001344	0.120536	4.435	6.786		
69	6/13/2012	5th bubble	29.999	5.3	7	0.001506	0.001336	0.127246	4.388	6.860		
70	6/13/2012	5th bubble	6.516	5.3	4	0.001506	0.001495	0.007358	4.388	1.508		
71	6/13/2012	5th bubble	4.137	5.3	1	0.001506	0.001510	-0.002649	4.388	0.966		

See Table A-1 notes for color coding explanation.

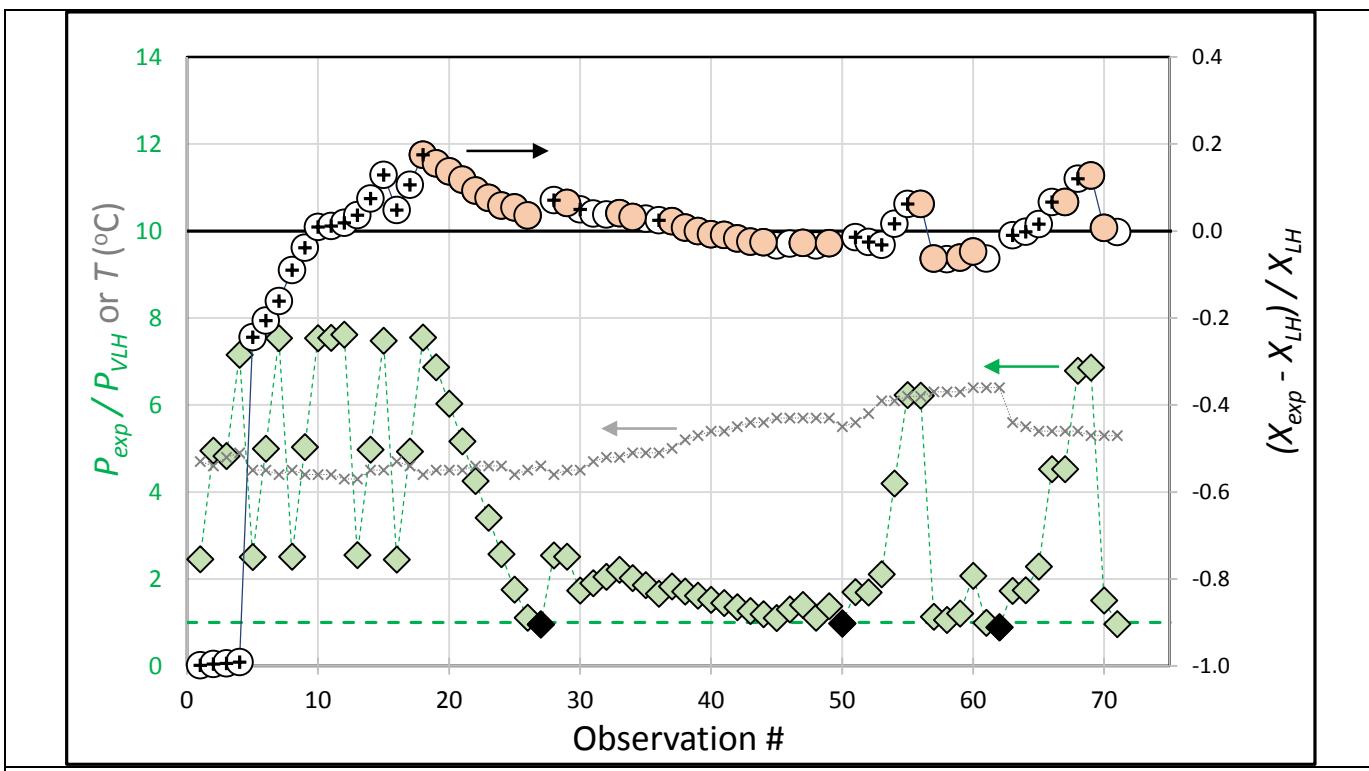


Figure A-2. Graphical representation of the sequence of observations in Data Set A-2 (CH4 in RO water). Symbol definitions provided in caption of Figure A-1.

Notes on Data Set A-2.

Gas additions were made to increase the dissolved CH₄ content after Observations #4, #7, #12, and #15. Hydrate was formed during observations of seven bubbles: #18, #28, #30, #36, #55, #66, and #68. The first gas exsolution (#27) did not appear to significantly eliminate a hydrate memory effect from the preceding bubble. The next two exsolutions were more effective, likely because the time of exsolution was increased and the system temperature was higher.

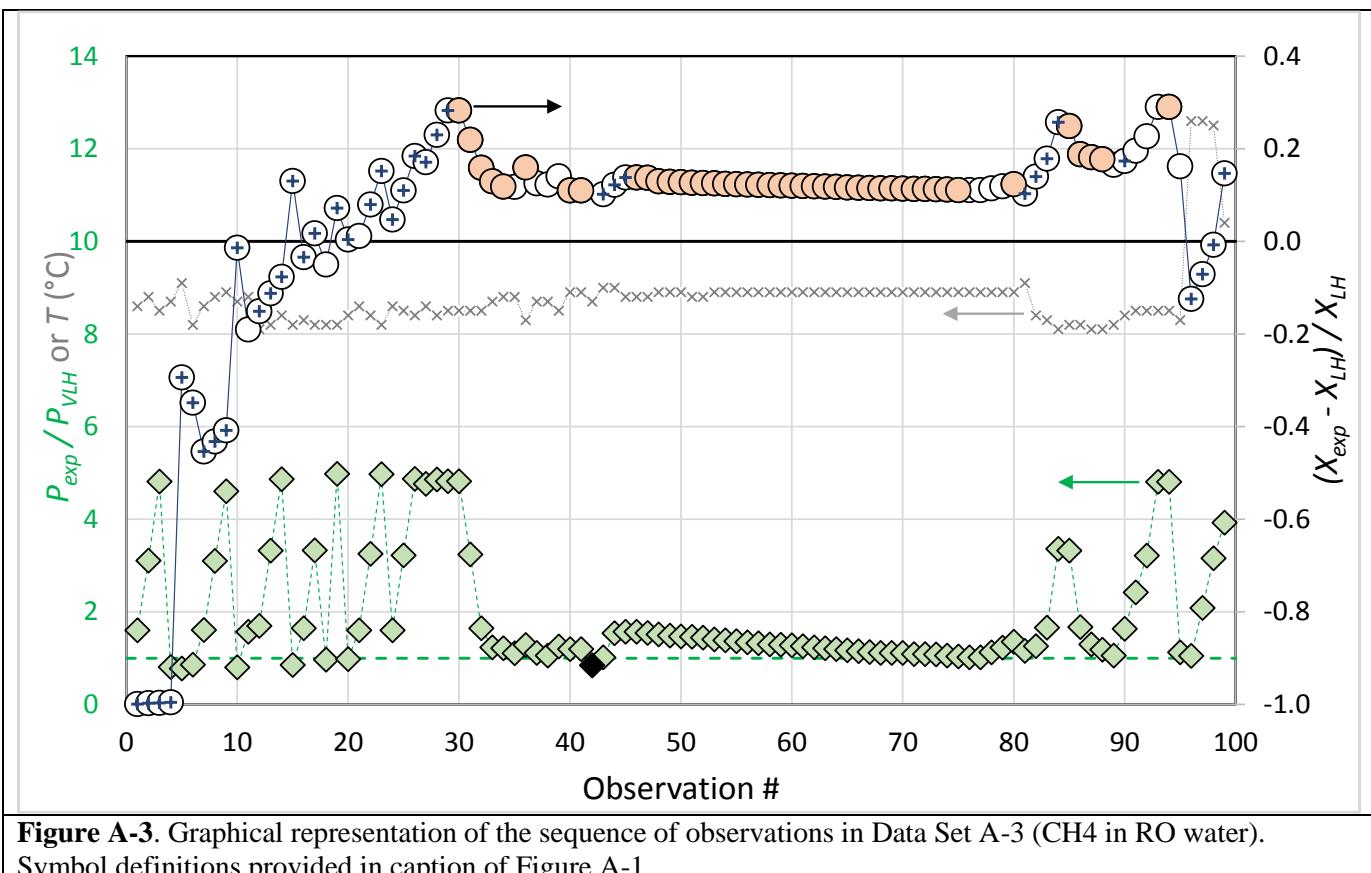
Table A-3. CH4 Data Set 3

Data Set A-3: CH4 in RO water. 8 to 13°C. Performed from 6/15/2012 to 07/6/2012.													
Observ. #	Sortable Date	Bubble #	Avg. P (MPa _{gauge})	Avg T (°C)	Observ. Time (min)	X _{exp} (mol frac.)	X _{LH} (mol frac.)	(X _{exp} - X _{LH})/X _{LH}	P _{VLH} @avg T (MPa _{absolute})	P _{avg} / P _{VLH}	(MPa _{absolute})	Dissolution rate (um/s)	
1	6/15/2012	1st bubble	9.997	8.6	23	0.000001	0.001815	-0.999449	6.317	1.599		1.5528	
2	6/15/2012	2nd bubble	19.967	8.8	41	0.000004	0.001747	-0.997711	6.457	3.108		1.0945	
3	6/18/2012	1st bubble	29.958	8.5	23	0.000005	0.001628	-0.996929	6.248	4.811		0.9806	
4	6/18/2012	2nd bubble	5.033	8.7	34	0.000007	0.001510	-0.995364	6.386	0.804		2.0178	
5	6/18/2012	3rd bubble	5.054	9.1	2	0.000989	0.001400	-0.293571	6.671	0.773			
6	6/19/2012	1st bubble	5.095	8.2	49	0.000990	0.001520	-0.348684	6.045	0.860			
7	6/19/2012	2nd bubble	10.039	8.6	38	0.000991	0.001815	-0.453994	6.317	1.605			
8	6/19/2012	3rd bubble	19.905	8.8	32	0.000992	0.001748	-0.432494	6.457	3.099			
9	6/19/2012	4th bubble	29.965	8.9	53	0.000989	0.001670	-0.407903	6.527	4.606		0.7202	
10	6/20/2012	1st bubble A	5.006	8.7	51	0.001489	0.001510	-0.013907	6.386	0.800		0.2052	
11	6/20/2012	1st bubble B	10.018	8.8	34	0.001489	0.001839	-0.190530	6.457	1.567		0.7012	
12	6/21/2012	1st bubble	10.032	8.1	44	0.001491	0.001757	-0.151191	5.979	1.695		0.8255	
13	6/21/2012	2nd bubble	19.995	8.2	6	0.001492	0.001681	-0.112433	6.045	3.324		1.0287	
14	6/21/2012	3rd bubble	29.951	8.4	28	0.001494	0.001618	-0.076625	6.180	4.863		0.6757	
15	6/22/2012	1st bubble	5.061	8.2	27	0.001707	0.001510	0.130464	6.045	0.854		-0.0457	
16	6/22/2012	2nd bubble	9.963	8.3	57	0.001719	0.001780	-0.034464	6.112	1.647		0.6219	
17	6/22/2012	3rd bubble	20.002	8.2	46	0.001710	0.001681	0.017477	6.045	3.326		0.647	
18	6/22/2012	3rd bubble	5.723	8.2	18	0.001710	0.001800	-0.050000	6.045	0.963		0.2021	
19	6/22/2012	4th bubble	29.979	8.2	27	0.001712	0.001597	0.071799	6.045	4.976		0.6338	
20	6/25/2012	1st bubble A	5.943	8.4	33	0.001836	0.001829	0.003827	6.180	0.978		0.1586	
21	6/25/2012	1st bubble B	10.004	8.6	37	0.001836	0.001815	0.011386	6.317	1.600		0.5752	
22	6/25/2012	2nd bubble	19.995	8.4	54	0.001838	0.001703	0.079272	6.180	3.252			
23	6/25/2012	3rd bubble	29.965	8.2	33	0.001840	0.001597	0.152160	6.045	4.974			
24	6/26/2012	1st bubble	10.018	8.6	72	0.001900	0.001815	0.046832	6.317	1.602		0.529	
25	6/26/2012	2nd bubble	20.002	8.5	48	0.001902	0.001713	0.110085	6.248	3.218		0.5857	
26	6/27/2012	1st bubble	29.985	8.4	48	0.001916	0.001618	0.184178	6.180	4.869			
27	6/27/2012	2nd bubble	29.992	8.6	47	0.001918	0.001638	0.171940	6.317	4.764			
28	6/28/2012	1st bubble	29.937	8.4	58	0.001990	0.001618	0.229843	6.180	4.861		0.5923	
29	6/28/2012	2nd bubble	30.006	8.5	5	0.002088	0.001628	0.282555	6.248	4.819			
30	6/28/2012	2nd bubble	30.006	8.5	16	0.002088	0.001628	0.282555	6.248	4.819			
31	6/28/2012	2nd bubble	20.140	8.5	12	0.002088	0.001713	0.218914	6.248	3.240			
32	6/28/2012	2nd bubble	10.170	8.5	18	0.002088	0.001802	0.158713	6.248	1.644			
33	6/28/2012	2nd bubble	7.770	8.7	23	0.002088	0.001848	0.129870	6.386	1.233		0.2718	
34	6/28/2012	2nd bubble	7.743	8.8		0.002088	0.001867	0.118372	6.457	1.215		-0.0531	
35	6/28/2012	2nd bubble	7.040	8.8	4	0.002088	0.001868	0.117773	6.457	1.106			
36	6/28/2012	2nd bubble	7.729	8.3	12	0.002088	0.001801	0.159356	6.112	1.281			
37	6/28/2012	2nd bubble	7.012	8.7	5	0.002088	0.001856	0.125000	6.386	1.114			
38	6/28/2012	2nd bubble	6.626	8.7	3	0.002088	0.001860	0.122581	6.386	1.053			
39	6/28/2012	2nd bubble	7.715	8.5	5	0.002088	0.001831	0.140360	6.248	1.251			
40	6/28/2012	2nd bubble	7.708	8.9	13	0.002088	0.001881	0.110048	6.527	1.196		0.177	
41	6/28/2012	2nd bubble	7.701	8.9	13	0.002088	0.001881	0.110048	6.527	1.195		0.0492	
42	6/29/2012	Exsolution	5.295	8.7	19				6.386	0.845			
43	6/29/2012	1st bubble	6.598	9	80	0.002090	0.001897	0.101740	6.599	1.015		0.2957	
44	6/29/2012	2nd bubble	10.032	9	3	0.002092	0.001864	0.122318	6.599	1.536			
45	6/29/2012	3rd bubble A	10.025	8.8	4	0.002094	0.001840	0.138043	6.457	1.568		0.9383	
46	6/29/2012	3rd bubble B	10.032	8.8	17	0.002094	0.001840	0.138043	6.457	1.569		0.2264	
47	6/29/2012	3rd bubble	9.880	8.8	1	0.002094	0.001841	0.137425	6.457	1.546			
48	6/29/2012	3rd bubble	9.749	8.9	1	0.002094	0.001854	0.129450	6.527	1.509			
49	6/29/2012	3rd bubble	9.604	8.9	1	0.002094	0.001856	0.128233	6.527	1.487			
50	6/29/2012	3rd bubble	9.460	8.9	1	0.002094	0.001857	0.127625	6.527	1.465			

Continued on next page.

Data Set A-3 continued: CH4 in RO water. 8 to 13°C. Performed from 6/15/2012 to 07/6/2012.												
Obsr. #	Sortable Date	Bubble #	Avg. P (MPa _{gauge})	Avg T (°C)	Observ. Time (min)	X _{exp} (mol frac.)	X _{LH} (mol frac.)	(X _{exp} - X _{LH}) / X _{LH}	P _{VLH} @ avg T (MPa _{absolute})	P _{avg} / P _{VLH} (MPa _{absolute})	Dissolution rate (um/s)	
51	6/29/2012	3rd bubble	9.322	8.8	1	0.002094	0.001859	0.126412	6.457	1.459		
52	6/29/2012	3rd bubble	9.191	8.8	1	0.002094	0.001860	0.125806	6.457	1.439		
53	6/29/2012	3rd bubble	9.060	8.9	1	0.002094	0.001861	0.125202	6.527	1.403		
54	6/29/2012	3rd bubble	8.929	8.9	1	0.002094	0.001863	0.123994	6.527	1.383		
55	6/29/2012	3rd bubble	8.798	8.9	1	0.002094	0.001864	0.123391	6.527	1.363		
56	6/29/2012	3rd bubble	8.639	8.9	1	0.002094	0.001865	0.122788	6.527	1.339		
57	6/29/2012	3rd bubble	8.515	8.9	1	0.002094	0.001866	0.122186	6.527	1.320		
58	6/29/2012	3rd bubble	8.377	8.9	1	0.002094	0.001867	0.121585	6.527	1.299		
59	6/29/2012	3rd bubble	8.322	8.9	1	0.002094	0.001868	0.120985	6.527	1.290		
60	6/29/2012	3rd bubble	8.246	8.9	1	0.002094	0.001869	0.120385	6.527	1.279		
61	6/29/2012	3rd bubble	8.094	8.9	1	0.002094	0.001870	0.119786	6.527	1.256		
62	6/29/2012	3rd bubble	7.936	8.9	1	0.002094	0.001872	0.118590	6.527	1.231		
63	6/29/2012	3rd bubble	7.826	8.9	1	0.002094	0.001873	0.117993	6.527	1.214		
64	6/29/2012	3rd bubble	7.688	8.9	1	0.002094	0.001874	0.117396	6.527	1.193		
65	6/29/2012	3rd bubble	7.529	8.9	1	0.002094	0.001876	0.116205	6.527	1.169		
66	6/29/2012	3rd bubble	7.412	8.9	1	0.002094	0.001877	0.115610	6.527	1.151		
67	6/29/2012	3rd bubble	7.288	8.9	1	0.002094	0.001878	0.115016	6.527	1.132		
68	6/29/2012	3rd bubble	7.164	8.9	1	0.002094	0.001879	0.114423	6.527	1.113		
69	6/29/2012	3rd bubble	7.164	8.9	7	0.002094	0.001879	0.114423	6.527	1.113		
70	6/29/2012	3rd bubble	7.143	8.9	2	0.002094	0.001880	0.113830	6.527	1.110		
71	6/29/2012	3rd bubble	7.005	8.9	2	0.002094	0.001881	0.113238	6.527	1.089		
72	6/29/2012	3rd bubble	7.005	8.9	1	0.002094	0.001881	0.113238	6.527	1.089		
73	6/29/2012	3rd bubble	6.936	8.9	1	0.002094	0.001882	0.112646	6.527	1.078		
74	6/29/2012	3rd bubble	6.798	8.9	1	0.002094	0.001883	0.112055	6.527	1.057		
75	6/29/2012	3rd bubble	6.640	8.9	1	0.002094	0.001885	0.110875	6.527	1.033		
76	6/29/2012	3rd bubble	6.522	8.9	2	0.002094	0.001886	0.110286	6.527	1.015		
77	6/29/2012	3rd bubble	6.522	8.9	1	0.002094	0.001886	0.110286	6.527	1.015		
78	6/29/2012	3rd bubble	7.219	8.9	4	0.002094	0.001879	0.114423	6.527	1.121		
79	6/29/2012	3rd bubble	7.908	8.9	5	0.002094	0.001872	0.118590	6.527	1.227		
80	6/29/2012	3rd bubble	8.715	8.9	1	0.002094	0.001864	0.123391	6.527	1.351		
81	7/2/2012	1st bubble	7.619	9.1	13	0.002096	0.001900	0.103158	6.671	1.157		
82	7/2/2012	2nd bubble	7.653	8.4	63	0.002067	0.001813	0.140099	6.180	1.255		
83	7/2/2012	3rd bubble	10.039	8.3	65	0.002098	0.001780	0.178652	6.112	1.659		
84	7/3/2012	1st bubble	19.988	8.1	6	0.002100	0.001670	0.257485	5.979	3.360	0.6071	
85	7/3/2012	1st bubble	19.988	8.2	79	0.002100	0.001681	0.249256	6.045	3.323		
86	7/3/2012	1st bubble	9.991	8.2	60	0.002100	0.001768	0.187783	6.045	1.669		
87	7/3/2012	1st bubble	7.660	8.1	74	0.002100	0.001777	0.181767	5.979	1.298		
88	7/3/2012	1st bubble	6.978	8.1	24	0.002100	0.001784	0.177130	5.979	1.184		
89	7/3/2012	1st bubble	6.288	8.2	5	0.002100	0.001802	0.165372	6.045	1.057		
90	7/3/2012	2nd bubble	10.004	8.4	23	0.002102	0.001792	0.173280	6.180	1.635	0.551	
91	7/3/2012	2nd bubble	15.010	8.5	11	0.002102	0.001758	0.195677	6.248	2.419	0.6203	
92	7/3/2012	2nd bubble	19.988	8.5	7	0.002102	0.001713	0.227087	6.248	3.215		
93	7/3/2012	2nd bubble	29.923	8.5	2	0.002102	0.001629	0.290362	6.248	4.806		
94	7/3/2012	2nd bubble	29.923	8.5	13	0.002102	0.001629	0.290362	6.248	4.806		
95	7/3/2012	2nd bubble	6.784	8.3	18	0.002102	0.001809	0.161968	6.112	1.127		
96	7/5/2012	1st bubble	9.997	12.6	75	0.002104	0.002402	-0.124166	9.636	1.048	0.4206	
97	7/5/2012	2nd bubble	19.988	12.6	54	0.002107	0.002267	-0.070760	9.636	2.085	0.5781	
98	7/5/2012	3rd bubble	29.985	12.5	53	0.002110	0.002126	-0.007616	9.537	3.155	0.5962	
99	7/6/2012	1st bubble	29.992	10.4	44	0.002112	0.001842	0.146748	7.665	3.926	0.5656	

See Table A-1 notes for color coding explanation.



Notes on Data Set A-3.

Gas additions were made to increase the dissolved CH₄ content after Observations #4, #9, #14, #19, #23, #27, and #28. Hydrate was formed during observations of four bubbles: #29, #45, #84, and #19. Observations #45 through #80 were used in Warzinski *et al.*, (2014). During #85 through #88 the hydrated bubble did not come into view in the HWTF. During #85 the bubble formed a hydrate shell and even had a hydrated wake. It went out of view after ~6 minutes and did not come back into view until the hydrate was decomposed by pressure reduction (#89).

Hydrate shedding was observed in #85 and #86. The bubble was allowed to remain out of view for an hour or more in #85, #86, and #87 to see if the hydrate would slowly decompose, which it did not appear to do.

Table A-4. CH4 Data Set 4

Data Set A-4: CH4 in RO water. 8 to 9°C. Performed from 10/15/2012 to 10/23/2012.											
Obsr. #	Sortable Date	Bubble #	Avg. P (MPa _{gauge})	Avg T (°C)	Observ. Time (min)	X _{exp} (mol frac.)	X _{LH} (mol frac.)	(X _{exp} - X _{LH})/ X _{LH}	P _{VLH} @avg T (MPa _{absolute})	P _{avg} / P _{VLH} (MPa _{absolute})	Dissolution rate (μm/s)
1	10/15/2012	1st bubble	29.937	8.4	27	0.000003	0.001618	-0.998146	6.180	4.861	0.9998
2	10/15/2012	2nd bubble	30.020	8.6	29	0.000007	0.001638	-0.995727	6.317	4.768	0.9637
3	10/17/2012	1st bubble	29.972	8.7	35	0.001493	0.001649	-0.094580	6.386	4.709	0.6783
4	10/17/2012	2nd bubble	29.979	8.8	48	0.001497	0.001660	-0.097928	6.457	4.659	0.6954
5	10/18/2012	Spray and bubble	6.536	8.3	-	0.001961	0.001812	0.082230	6.112	1.086	
6	10/18/2012	1st bubble	29.792	8.9	0.5	0.001961	0.001672	0.172847	6.527	4.580	
7	10/18/2012	2nd bubble	29.965	9.1	52	0.001966	0.001692	0.161939	6.671	4.507	0.6073
8	10/19/2012	1st bubble	29.985	8.7	64	0.002051	0.001649	0.243904	6.386	4.711	0.5898
9	10/22/2012	1st bubble	29.979	8.6	65	0.002145	0.001638	0.309222	6.317	4.762	0.5811
10	10/22/2012	2nd bubble	29.985	8.9	33	0.002149	0.001670	0.286708	6.527	4.609	0.5752
11	10/22/2012	Upflow to mix	29.992	8.9	19	0.002149	0.001671	0.286056	6.527	4.610	
12	10/22/2012	Exsolution	5.309	8.4	68				6.180	0.876	
13	10/23/2012	1st bubble	22.512	8.2	0.6	0.002155	0.001659	0.298975	6.045	3.741	
14	10/23/2012	1st bubble	23.001	8.2	5	0.002155	0.001655	0.302115	6.045	3.822	
15	10/23/2012	1st bubble	7.701	8.6	10	0.002155	0.001837	0.173108	6.317	1.235	
16	10/23/2012	Exsolution	4.950	8.1	32				5.979	0.845	
17	10/23/2012	2nd bubble	9.977	8.4	3	0.002159	0.001792	0.204799	6.180	1.631	
18	10/23/2012	2nd bubble	9.977	8.4	15	0.002159	0.001792	0.204799	6.180	1.631	-0.4024
19	10/23/2012	2nd bubble	7.584	8.4	11	0.002159	0.001814	0.190187	6.180	1.244	
20	10/23/2012	2nd bubble	6.171	8.3	4	0.002159	0.001815	0.189532	6.112	1.026	
21	10/23/2012	3rd bubble	6.171	8.3	1	0.002161	0.001815	0.190634	6.112	1.026	
22	10/23/2012	3rd bubble	6.840	8.44	1	0.002161	0.001821	0.186711	6.207	1.118	
23	10/23/2012	3rd bubble	7.653	8.47	1	0.002161	0.001825	0.184110	6.227	1.245	
24	10/23/2012	3rd bubble	8.191	8.50	3	0.002161	0.001820	0.187363	6.248	1.327	
25	10/23/2012	3rd bubble	8.879	8.51	2	0.002161	0.001814	0.191290	6.255	1.436	
26	10/23/2012	3rd bubble	9.702	8.53	2	0.002161	0.001806	0.196567	6.269	1.564	
27	10/23/2012	3rd bubble	10.258	8.54	2	0.002161	0.001801	0.199889	6.275	1.651	
28	10/23/2012	3rd bubble	11.613	8.56	2	0.002161	0.001801	0.199889	6.289	1.863	
29	10/23/2012	3rd bubble	13.609	8.57	2	0.002161	0.001782	0.212682	6.296	2.178	
30	10/23/2012	3rd bubble	17.167	8.59	3	0.002161	0.001750	0.234857	6.310	2.737	
31	10/23/2012	3rd bubble	20.596	8.58	3	0.002161	0.001719	0.257126	6.303	3.284	
32	10/23/2012	3rd bubble	24.181	8.58	3	0.002161	0.001688	0.280213	6.303	3.853	
33	10/23/2012	3rd bubble	7.715	8.6	31	0.002161	0.001837	0.176375	6.317	1.237	0.2301
34	10/23/2012	3rd bubble	7.005	8.68	2	0.002161	0.001856	0.164332	6.372	1.115	
35	10/23/2012	3rd bubble	7.611	8.70	3	0.002161	0.001850	0.168108	6.386	1.208	
36	10/23/2012	3rd bubble	8.349	8.7	6	0.002161	0.001843	0.172545	6.386	1.323	
37	10/23/2012	3rd bubble	7.584	8.6	0.3	0.002161	0.001838	0.175734	6.317	1.217	

See Table A-1 notes for color coding explanation.

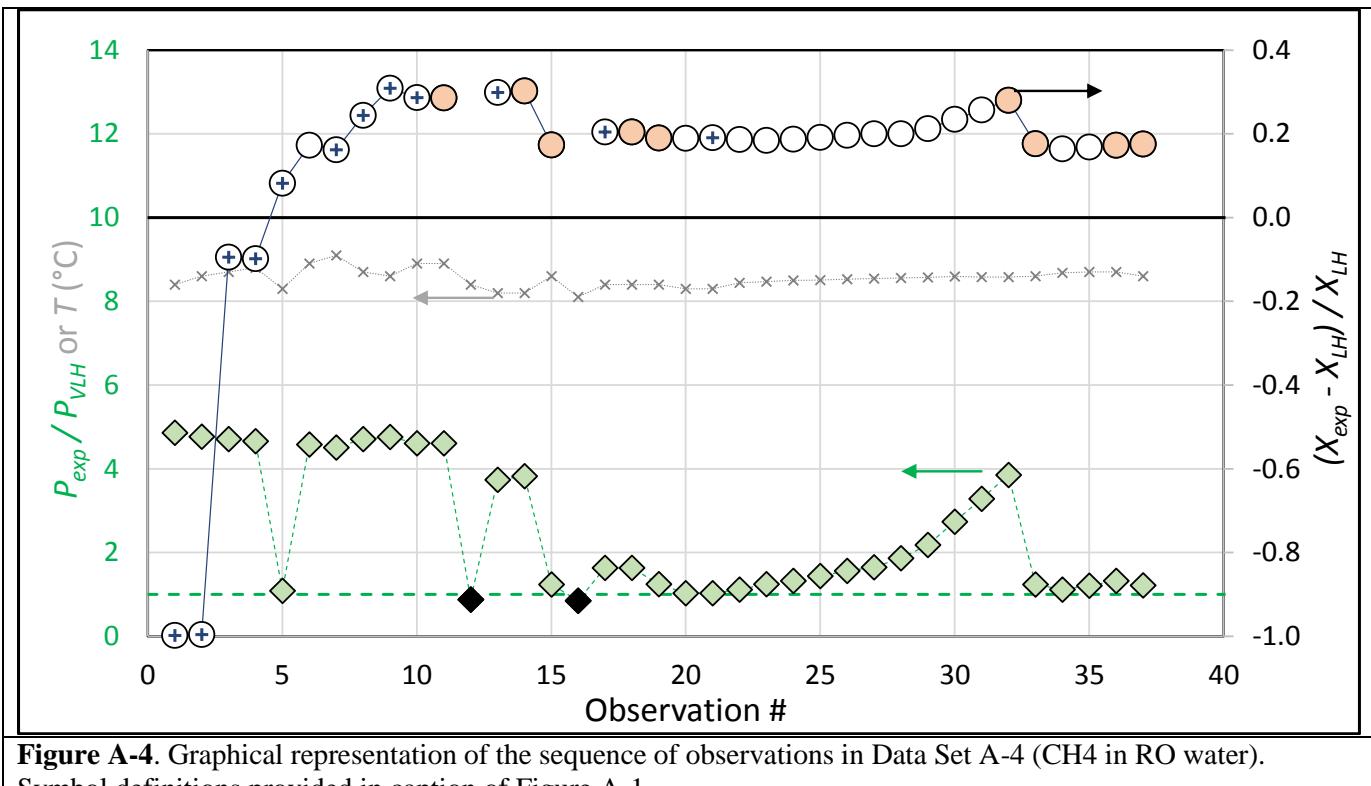


Figure A-4. Graphical representation of the sequence of observations in Data Set A-4 (CH₄ in RO water). Symbol definitions provided in caption of Figure A-1.

Notes on Data Set A-4.

Gas additions were made to increase the dissolved CH₄ content after Observations #2, #4, #7, and #8. Observation #5 consistent of a short spray of CH₄ into the HWTF that was just inside the VLH equilibrium region and also slightly supersaturated. It did not form hydrate. At the end of the observation of the 2nd bubble on 10/22/2012 (#10) the HWTF was put into upflow to mix. Hydrate formed as evidenced by a small, misshapen, hydrate-covered bubble that was somewhat stable (#11). After hydrate formed on the next two bubbles on 10/23/2012, the pressure was lowered to decompose the hydrate after the first bubble (#16); however, after hydrate formed on the second bubble, the HWTF was put into upflow to mix the unit while going close to, but not below, the VLH equilibrium (#20). The goal was to not eliminate the memory effect by exsolution, which did not happen, but to quickly inject the next bubble (#21) and then steadily, but quickly (1 to 2 minute intervals) raise the pressure as indicated in Table A-4, #22 to #32, where hydrate formed on the bubble (#32).

Table A-5. CH4 Data Set 5

Data Set A-5: CH4 in RO water. 1 to 3°C. Performed from 10/25/2012 to 11/02/2012.											
Obsr. #	Sortable Date	Bubble #	Avg. P (MPa _{gauge})	Avg T (°C)	Observ. Time (min)	X _{exp} (mol frac.)	X _{LH} (mol frac.)	(X _{exp} - X _{LH})/X _{LH}	P _{V LH} @avg T (MPa _{absolute})	P _{avg} / P _{V LH} (MPa _{absolute})	Dissolution rate (um/s)
1	10/25/2012	1st bubble	29.985	2.7	29	0.000004	0.001146	-0.996510	3.383	8.892	0.8963
2	10/25/2012	2nd bubble	9.997	3	17	0.000005	0.001279	-0.996089	3.479	2.902	1.4127
3	10/25/2012	3rd bubble	19.988	2.9	22	0.000008	0.001214	-0.993411	3.447	5.828	0.9728
4	10/29/2012	1st bubble	29.972	1.8	33	0.000586	0.001088	-0.461462	3.116	9.652	0.7704
5	10/29/2012	2nd bubble	29.999	2.1	31	0.000589	0.001107	-0.467880	3.202	9.399	0.7709
6	10/30/2012	1st bubble	29.923	1.8	4	0.000729	0.001088	-0.329963	3.116	9.636	
7	10/30/2012	2nd bubble	29.992	2	28	0.000733	0.001101	-0.333990	3.173	9.483	0.7427
8	10/31/2012	1st bubble	29.972	1.6	39	0.000999	0.001076	-0.071355	3.059	9.831	0.7118
9	10/31/2012	2nd bubble	30.006	1.6	23	0.001003	0.001076	-0.067490	3.059	9.843	0.7082
10	10/31/2012	3rd bubble	29.930	1.6	28	0.001127	0.001076	0.047432	3.059	9.818	0.6889
11	10/31/2012	4th bubble	29.979	1.7	32	0.001131	0.001082	0.045387	3.087	9.743	0.6863
12	11/1/2012	~12 bubbles	2.910	1.4	0.3	0.001205	0.001200	0.004167	3.002	1.003	
13	11/1/2012	1st bubble	26.200	1.5	2	0.001209	0.001088	0.111213	3.031	8.679	
14	11/1/2012	1st bubble	28.751	1.5	7	0.001209	0.001075	0.124651	3.031	9.520	
15	11/1/2012	1st bubble	10.066	1.5	3	0.001209	0.001170	0.033333	3.031	3.355	
16	11/1/2012	1st bubble	3.640	1.5	10	0.001209	0.001204	0.004153	3.031	1.235	
17	11/1/2012	1st bubble	3.144	1.5	3	0.001209	0.001206	0.002488	3.031	1.071	
18	11/1/2012	Exsolution	2.551	1.5	35				3.031	0.875	
19	11/1/2012	2nd bubble A	4.992	2	27	0.001211	0.001232	-0.017045	3.173	1.605	0.7024
20	11/1/2012	2nd bubble B	9.956	2	14	0.001211	0.001205	0.004761	3.173	3.169	0.896
21	11/1/2012	3rd bubble	29.972	1.9	29	0.001214	0.001094	0.109296	3.144	9.564	0.6892
22	11/2/2012	1st bubble	29.972	1.4	8	0.001218	0.001064	0.145173	3.002	10.016	0.7282
23	11/2/2012	1st bubble	29.958	1.4	5	0.001218	0.001063	0.145814	3.002	10.012	
24	11/2/2012	1st bubble	20.064	1.4	5	0.001218	0.001112	0.095324	3.002	6.716	
25	11/2/2012	1st bubble	10.191	1.3	6	0.001218	0.001156	0.053633	2.974	3.460	
26	11/2/2012	1st bubble	4.826	1.3	22	0.001218	0.001183	0.029586	2.974	1.657	
27	11/2/2012	1st bubble	4.275	1.4	25	0.001218	0.001193	0.020956	3.002	1.458	
28	11/2/2012	1st bubble	3.654	1.4	37	0.001218	0.001196	0.018395	3.002	1.251	
29	11/2/2012	1st bubble	2.979	1.5	11	0.001218	0.001207	0.009114	3.031	1.016	
30	11/2/2012	Exsolution	2.627	1.5	5				3.031	0.900	

See Table A-1 notes for color coding explanation.

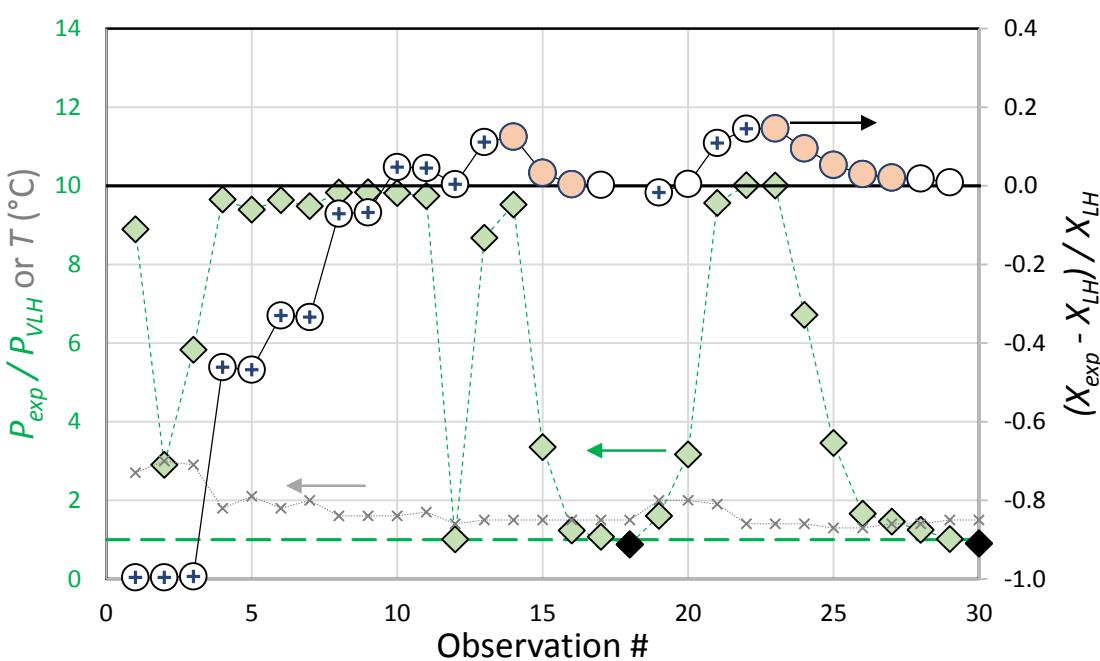


Figure A-5. Graphical representation of the sequence of observations in Data Set A-5 (CH4 in RO water). Symbol definitions provided in caption of Figure A-1.

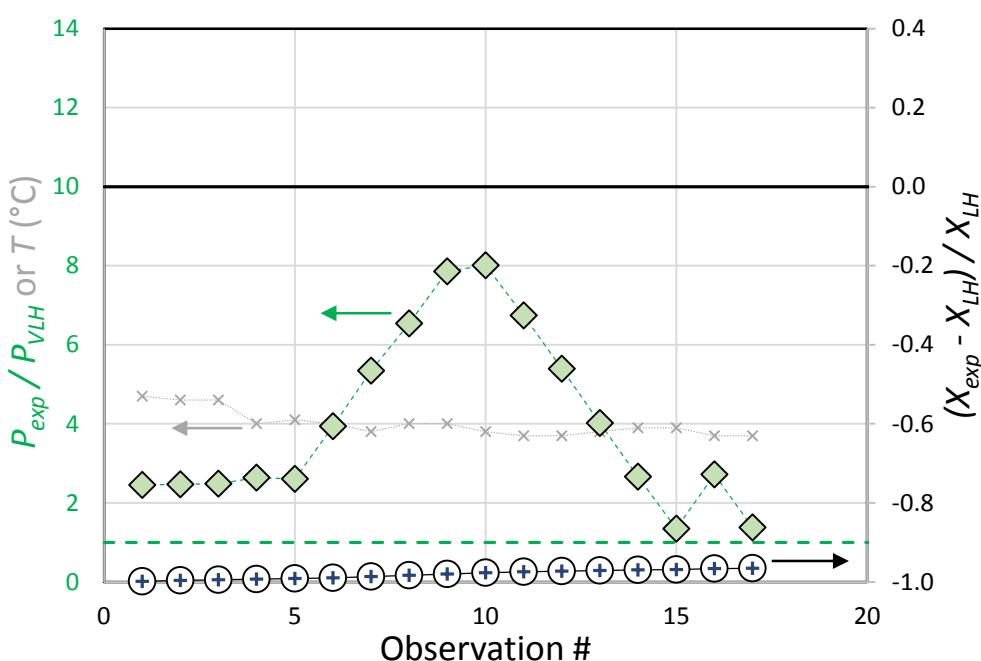
Notes on Data Set A-5.

Gas additions were made to increase the dissolved CH4 content after Observations #3, #5, #7, and #9. Observation #12 involved ~12 small bubbles released from the injection port while the HWTF was in upflow, so they circulated through the pump and flow loop. The system was just at VLH equilibrium ($P_{exp}/P_{VLH} = 1$) and at LH saturation ($(X_{exp} - X_{LH})/X_{LH} = 0$). Hydrate was not formed on these small bubbles at these conditions. However, hydrate formed on the next bubble, initially as ring like structures that may indicate that as P was being increased during the release of the bubble in #13 some of the small circulating bubbles from #12 formed hydrate or hydrate particles that initiated hydrate formation when they impacted the bubble in #13. Gas exsolution (#18) was effective in eliminating any hydrate memory effect as evidenced by the next two bubbles (#19, #20, and #21). The first two of these was the same bubble with a pressure increase during observation.) Even the next bubble (1st bubble on 11/2/2012) did not form hydrate until after 8 minutes.

Table A-6. CH4 Data Set 6

Data Set A-6: CH4 in RO water. 4 to 5°C. Performed from 6/24/2014 to 7/3/2014.											
Observer #	Sortable Date	Bubble #	Avg. P (MPa _{gauge})	Avg T (°C)	Observ. Time (min)	X _{exp} (mol frac.)	X _{LH} (mol frac.)	(X _{exp} - X _{LH})/X _{LH}	P _{VLH} @ avg T (MPa _{absolute})	P _{avg} / P _{VLH} (MPa _{absolute})	Dissolution rate (um/s)
1	6/24/2014	Bubble #1	9.997	4.7	26	0.000002	0.001417	-0.998589	4.118	2.453	1.2994
2	6/24/2014	Bubble #2	9.942	4.6	22	0.000005	0.001409	-0.996451	4.075	2.465	1.2768
3	6/24/2014	Bubble #3	10.025	4.6	20	0.000007	0.001408	-0.995028	4.075	2.485	1.3431
4	6/25/2014	Bubble #1	10.018	4	25	0.000009	0.001358	-0.993373	3.833	2.640	1.2977
5	6/25/2014	Bubble #2	9.991	4.1	25	0.000012	0.001366	-0.991215	3.872	2.607	1.3721
6	6/26/2014	Bubble #1	14.989	4	34	0.000014	0.001327	-0.989450	3.833	3.937	1.0682
7	6/26/2014	Bubble #2	19.995	3.8	26	0.000017	0.001281	-0.986729	3.758	5.348	0.9403
8	6/26/2014	Bubble #3	24.966	4	33	0.000021	0.001266	-0.983412	3.833	6.540	0.9121
9	6/26/2014	Bubble #4	30.013	4	31	0.000025	0.001236	-0.979773	3.833	7.856	0.8422
10	6/27/2014	Bubble #1	29.999	3.8	36	0.000028	0.001222	-0.977087	3.758	8.010	0.8585
11	6/27/2014	Bubble #2	25.001	3.7	29	0.000032	0.001244	-0.974277	3.721	6.746	0.871
12	7/2/2014	Bubble #1	19.981	3.7	35	0.000035	0.001273	-0.972506	3.721	5.397	0.9617
13	7/2/2014	Bubble #2	15.010	3.8	21	0.000038	0.001311	-0.971014	3.758	4.021	1.0743
14	7/2/2014	Bubble #3	10.018	3.9	23	0.000041	0.001350	-0.969630	3.795	2.666	1.3775
15	7/2/2014	Bubble #4	5.013	3.9	29	0.000043	0.001381	-0.968863	3.795	1.347	1.8221
16	7/3/2014	Bubble #1	10.018	3.7	24	0.000045	0.001333	-0.966242	3.721	2.720	1.3336
17	7/3/2014	Bubble #2	5.013	3.7	2	0.000047	0.001365	-0.965568	3.721	1.374	

See Table A-1 notes for color coding explanation.

**Figure A-6.** Graphical representation of the sequence of observations in Data Set A-6 (CH4 in RO water). Symbol definitions provided in caption of Figure A-1.**Notes on Data Set A-5.**

No gas additions were made. The purpose of this set of experiments was to obtain dissolution rates from multiple observations at different pressures.

Table A-7. CH4 Data Set 6

Data Set A-7: CH4 in RO water. 2 to 10°C. Performed from 7/9/2014 to 7/31/2014.											
Obsr. #	Sortable Date	Bubble #	Avg. P (MPa _{gauge})	Avg T (°C)	Observ. Time (min)	X _{exp} (mol frac.)	X _{LH} (mol frac.)	(X _{exp} - X _{LH})/X _{LH}	P _{VLH} @avg T (MPa _{absolute})	P _{avg} / P _{VLH} (MPa _{absolute})	Dissolution rate (um/s)
1	7/9/2014	1st bubble	10.011	4.1	38	0.000003	0.001248	-0.997692	4.499	2.248	1.029
2	7/9/2014	2nd bubble	10.004	4.2	41	0.000006	0.001256	-0.995525	4.545	2.224	1.0199
3	7/9/2014	3rd bubble	10.004	3.8	33	0.000008	0.001225	-0.993241	4.364	2.316	1.0146
4	7/10/2014	1st bubble	20.002	3.6	42	0.000012	0.001154	-0.989775	4.276	4.701	0.695
5	7/10/2014	2nd bubble	20.009	3.7	33	0.000016	0.001161	-0.986649	4.320	4.655	0.7009
6	7/10/2014	3rd bubble	29.992	3.7	61	0.000020	0.001107	-0.982204	4.320	6.967	0.6468
7	7/11/2014	1st bubble	29.999	3.6	60	0.000024	0.001100	-0.978182	4.276	7.039	0.6395
8	7/11/2014	2nd bubble	29.999	3.7	41	0.000029	0.001107	-0.973803	4.320	6.968	0.6434
9	7/16/2014	1st bubble	10.004	3.2	40	0.000031	0.001181	-0.973751	4.107	2.461	0.9902
10	7/16/2014	2nd bubble	10.004	2.3	45	0.000034	0.001119	-0.969616	3.754	2.692	0.9765
11	7/17/2014	1st bubble	20.009	2.4	59	0.000038	0.001075	-0.964651	3.792	5.304	0.6775
12	7/17/2014	2nd bubble	20.002	2	55	0.000041	0.001050	-0.960952	3.646	5.514	0.6761
13	7/18/2014	1st bubble	29.992	2.3	59	0.000045	0.001020	-0.955882	3.754	8.016	0.6269
14	7/18/2014	2nd bubble	29.999	2.1	67	0.000050	0.001008	-0.950397	3.681	8.176	0.624
15	7/23/2014	1st bubble	29.910	2.8	11	0.001145	0.001050	0.090476	3.945	7.607	
16	7/23/2014	1st bubble	19.995	2.8	12	0.001145	0.001100	0.040909	3.945	5.094	
17	7/23/2014	1st bubble	10.018	2.8	10	0.001145	0.001153	-0.006938	3.945	2.565	
18	7/23/2014	1st bubble	5.019	3.1	32	0.001145	0.001202	-0.047421	4.066	1.259	0.2365
19	7/23/2014	1st bubble	4.509	3	9	0.001145	0.001197	-0.043442	4.025	1.145	
20	7/23/2014	Exsolution	3.689	3	43				4.025	0.942	
21	7/23/2014	2nd bubble	9.791	2.7	0.5	0.001148	0.001147	0.000872	3.906	2.532	
22	7/23/2014	2nd bubble	9.846	2.8	15	0.001148	0.001154	-0.005199	3.945	2.521	
23	7/23/2014	2nd bubble	4.964	2.8	7	0.001148	0.001180	-0.027119	3.945	1.284	
24	7/23/2014	2nd bubble	4.737	2.8	9	0.001148	0.001182	-0.028765	3.945	1.226	
25	7/23/2014	2nd bubble	4.461	2.8	23	0.001148	0.001183	-0.029586	3.945	1.156	0.1285
26	7/23/2014	2nd bubble	4.447	3.1	35	0.001148	0.001205	-0.047303	4.066	1.119	0.1815
27	7/23/2014	2nd bubble	4.999	3.2	7	0.001148	0.001209	-0.050455	4.107	1.242	
28	7/23/2014	2nd bubble	10.246	3.2	3	0.001148	0.001180	-0.027119	4.107	2.519	
29	7/23/2014	2nd bubble	4.978	3.2	7	0.001148	0.001209	-0.050455	4.107	1.237	
30	7/23/2014	Exsolution	3.392	3.1	82				4.066	0.859	
31	7/24/2014	1st bubble	29.992	5.8	85	0.001153	0.001257	-0.082737	5.351	5.624	0.4851
32	7/24/2014	2nd bubble	30.020	5.8	13	0.001158	0.001257	-0.078759	5.351	5.630	0.4587
33	7/24/2014	2nd bubble	29.992	5.6	34	0.001158	0.001242	-0.067633	5.242	5.741	0.4455
34	7/24/2014	2nd bubble	29.979	5.6	15	0.001158	0.001242	-0.067633	5.242	5.738	
35	7/24/2014	2nd bubble	5.936	5.8	15	0.001158	0.001417	-0.182781	5.351	1.128	0.5969
36	7/24/2014	2nd bubble	5.902	5.7	18	0.001158	0.001408	-0.177557	5.296	1.134	0.4002
37	7/24/2014	2nd bubble	5.881	5.8	22	0.001158	0.001417	-0.182781	5.351	1.118	
38	7/24/2014	Exsolution	3.447	5.3	42				5.084	0.698	
39	7/25/2014	1st bubble	30.006	6.1	49	0.001164	0.001281	-0.091335	5.517	5.457	0.4838
40	7/25/2014	1st bubble	9.991	6.1	27	0.001164	0.001415	-0.177385	5.517	1.829	0.5429
41	7/25/2014	2nd bubble	29.999	6.1	34	0.001226	0.001281	-0.042935	5.517	5.456	0.4839
42	7/28/2014	1st bubble	30.006	6.1	69	0.001325	0.001281	0.034348	5.517	5.457	0.4538
43	7/28/2014	2nd bubble	30.020	6.1	29	0.001420	0.001281	0.108509	5.517	5.459	0.4465
44	7/29/2014	1st bubble	30.006	6.2	1	0.001515	0.001289	0.175330	5.574	5.401	
45	7/29/2014	1st bubble	30.006	6.2	22	0.001515	0.001289	0.175330	5.574	5.401	
46	7/29/2014	1st bubble	9.991	6.2	5	0.001515	0.001424	0.063904	5.574	1.810	
47	7/29/2014	1st bubble	8.232	6.2	7	0.001515	0.001437	0.054280	5.574	1.495	
48	7/29/2014	1st bubble	6.012	6.2	14	0.001515	0.001453	0.042670	5.574	1.097	
49	7/29/2014	Exsolution & heating	5.323	7.3	63				6.242	0.869	
50	7/29/2014	2nd bubble	29.799	6.1	10	0.001522	0.001282	0.187207	5.517	5.419	

Continued on next page.

Data Set A-7 continued: CH4 in RO water. 2 to 10°C. Performed from 7/9/2014 to 7/31/2014.													
Obser. #	Sortable Date	Bubble #	Avg. P (MPa _{gauge})	Avg T (°C)	Observ. Time (min)	X _{exp} (mol frac.)	X _{LH} (mol frac.)	(X _{exp} - X _{LH})/X _{LH}	P _{VLH} @ avg T (MPa _{absolute})	P _{avg} / P _{VLH} (MPa _{absolute})	Dissolution rate (um/s)		
51	7/29/2014	2nd bubble	8.005	6.1	22	0.001522	0.001429	0.065080	5.517	1.469	0.5696		
52	7/29/2014	2nd bubble	6.950	6.3	20	0.001522	0.001455	0.046048	5.632	1.252			
53	7/29/2014	2nd bubble	6.391	6.2	11	0.001522	0.001450	0.049655	5.574	1.165			
54	7/29/2014	2nd bubble	6.033	6.2	30	0.001522	0.001453	0.047488	5.574	1.100			
55	7/29/2014	Exsolution & heating	5.585	8.2	61				6.855	0.829			
56	7/30/2014	1st bubble	30.006	8.2	2	0.001528	0.001463	0.044429	6.855	4.392			
57	7/30/2014	1st bubble	30.013	8.2	15	0.001528	0.001463	0.044429	6.855	4.393			
58	7/30/2014	1st bubble	7.102	8.1	5	0.001528	0.001638	-0.067155	6.784	1.062			
59	7/30/2014	1st bubble	7.102	8.1	5	0.001528	0.001638	-0.067155	6.784	1.062			
60	7/30/2014	Exsolution	5.102	8.2	73				6.855	0.759			
61	7/30/2014	2nd bubble	10.018	8.2	15	0.001533	0.001625	-0.056615	6.855	1.476			
62	7/30/2014	2nd bubble	9.991	8.1	44	0.001533	0.001614	-0.050186	6.784	1.488	0.4093		
63	7/30/2014	3rd bubble	19.995	8.3	3	0.001538	0.001552	-0.009021	6.927	2.901			
64	7/30/2014	3rd bubble	19.967	8.2	6	0.001538	0.001542	-0.002594	6.855	2.928			
65	7/30/2014	3rd bubble	10.011	8.3	34	0.001538	0.001636	-0.059902	6.927	1.460	0.2415		
66	7/30/2014	3rd bubble	9.294	8.2	10	0.001538	0.001631	-0.057020	6.855	1.371			
67	7/30/2014	3rd bubble	8.612	8.1	7	0.001538	0.001626	-0.054121	6.784	1.284			
68	7/30/2014	3rd bubble	8.260	8	5	0.001538	0.001617	-0.048856	6.713	1.245			
69	7/30/2014	3rd bubble	7.908	7.9	5	0.001538	0.001610	-0.044720	6.644	1.206			
70	7/30/2014	3rd bubble	7.557	8	4	0.001538	0.001623	-0.052372	6.713	1.141			
71	7/30/2014	3rd bubble	7.205	8	4	0.001538	0.001627	-0.054702	6.713	1.088			
72	7/30/2014	3rd bubble	6.888	8.1	16	0.001538	0.001640	-0.062195	6.784	1.030			
73	7/30/2014	3rd bubble	8.253	8.2	1	0.001538	0.001639	-0.061623	6.855	1.219			
74	7/30/2014	3rd bubble	8.598	8.2	5	0.001538	0.001637	-0.060476	6.855	1.269			
75	7/30/2014	3rd bubble	7.088	8.3	4	0.001538	0.001661	-0.074052	6.927	1.038			
76	7/30/2014	3rd bubble	8.577	8.3	7	0.001538	0.001648	-0.066748	6.927	1.253			
77	7/30/2014	3rd bubble	7.481	8.3	2	0.001538	0.001657	-0.071817	6.927	1.095			
78	7/30/2014	3rd bubble	9.280	8.3	2	0.001538	0.001642	-0.063337	6.927	1.354			
79	7/30/2014	3rd bubble	7.922	8.3	14	0.001538	0.001654	-0.070133	6.927	1.158			
80	7/31/2014	1st bubble	29.786	10.3	2	0.001545	0.001685	-0.083086	8.586	3.481			
81	7/31/2014	1st bubble	29.806	10.3	11	0.001545	0.001685	-0.083086	8.586	3.483			
82	7/31/2014	1st bubble	10.018	10.4	26	0.001545	0.001894	-0.184266	8.681	1.166	0.2132		
83	7/31/2014	1st bubble	8.274	10.0	14	0.001545	0.001858	-0.168461	8.308	1.008			

See Table A-1 notes for color coding explanation.

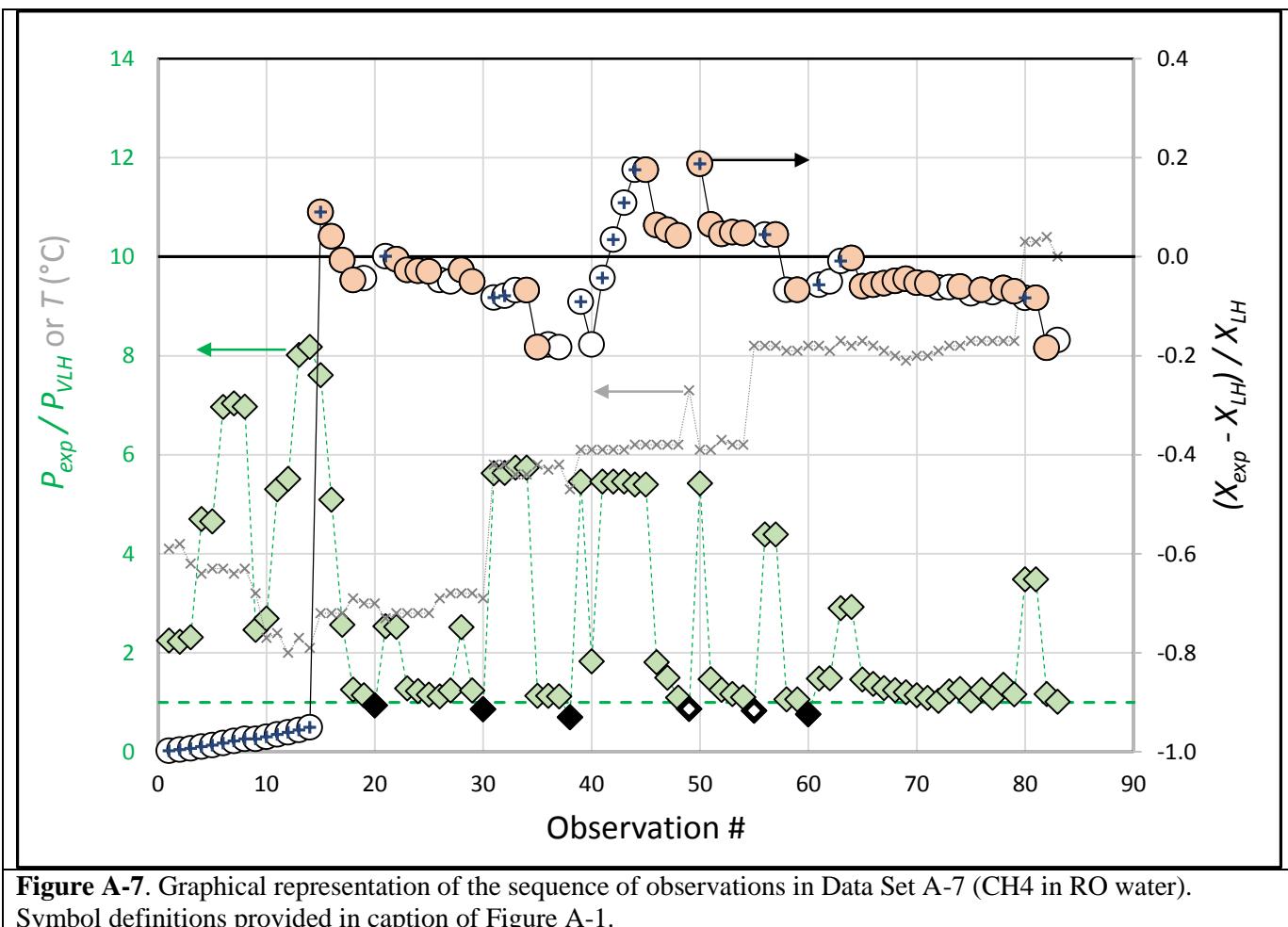


Figure A-7. Graphical representation of the sequence of observations in Data Set A-7 (CH₄ in RO water). Symbol definitions provided in caption of Figure A-1.

Notes on Data Set A-7.

Gas additions were made to increase the dissolved CH₄ content after Observations #14, #40, #41, #42, and #43. Hydrate first formed on the bubble in Observation #15 after it was released from the bubble cup in the HWTF at ~20.7 MPa and while pressure was being increased to ~30.0 MPa. Hydrate formed rapidly even after gas exsolution (#20) on the next bubble (#21). Increasing the temperature in the HWTF after the exsolution at #30 enabled the next bubble (#31) to be observed without hydrate formation at ~30 MPa for 85 minutes. The next bubble (#32) also did not form hydrate for 57 minutes and was much smaller when it did. The next gas exsolution (#38), the first at a higher T , effectively eliminated any hydrate memory effect, as evidenced in #40. The new bubbles at Observations #41, #42, #43, and #44 were all observed at ~30 MPa after small additions of gas were made to increase the gas saturation before each bubble. Hydrate did not form until #44; at which point the system was ~18% supersaturated in dissolved gas content. Additional bubbles were then observed, even at higher temperatures; however the hydrate memory effect could not be completely eliminated, except at lower pressures (see #61 and #62. #62 is a smaller part that shed from #61, which was a large bubble).